Title: Feasibility Study for a Demonstration Plant for Liquefaction and

Coprocessing of Waste Plastics and Tires

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Introduction

Since 1993, a research program on the liquefaction of waste polymers and the coprocessing of waste polymers with coal has been sponsored by the U.S. Department of Energy. The research has been carried out by academic, industrial and government scientists and engineers. Much of the laboratory research has been conducted by the Consortium for Fossil Fuel Liquefaction Science (CFFLS), a five university research consortium with participants from the University of Kentucky, Auburn University, the University of Pittsburgh, the University of Utah and West Virginia University. Industrial participation has been provided by Hydrocarbon Technologies, Inc. (HTI), where pilot scale and continuous tests have conducted, Consol, where specialized analytical techniques have been employed, and the Mitre Corporation, where economic analyses have been carried out. Research at the Federal Energy Technology Center (FETC, Pittsburgh), has complemented work in the academic and industrial sectors.

The current paper presents a brief summary of a feasibility study for a demonstration plant for this technology. A complete report of this study ⁽¹⁾ has been prepared. The study was conducted by a committee that included participants from CFFLS, FETC and Burns & Roe, listed below.

<u>CFFLS</u>		<u>FETC</u>	Burns & Roe	
G.P. Huffman	J. Zondlo	M. Eastman	H. McIlvried	
L.L. Anderson	I. Wender	M. Baird	H. Schindler	
M. El-Halwagi	J. Tierney	A. Cugini	J. Wilbur	
N. Shah		S. Lee	R. Srivastava	
M. Shelley		U. Rao		
A.R. Tarrer		K. Rothenberger		

The primary goals of the study were to establish a conceptual design for the plant, to carry out an economic analysis, and to develop a group of stakeholders for the technology.

Potential Resource and Current Practice in the U.S.

Over 44 billion lbs. of waste plastic are disposed of in the U.S. each year, which is 175 lbs. for every person in the country. Approximately 50% of PET soft drink bottles and 30% of HDPE milk and water bottles are mechanically recycled (melted and re-extruded) but recycling of other types of waste plastic is very limited. Only about 5% of all waste plastic produced in the U.S. is currently recycled.

Similarly, the recycling of the 300 million waste tires generated annually in the U.S. is very limited. (1) Crumb rubber from used tires is used to a limited extent as an additive for new tires, as an additive to asphalt, and for the production of mats used for playgrounds, running tracks and stables. Only about 15% of used tires are currently recycled in this country. The use of tires as tire-derived fuel (TDF) is significant (4) but it is not recycling. Approximately 75% of the used tires in this country end up in landfills, stockpiles or illegal dumps.

The U.S. generates approximately 30 million barrels of waste oils, greases, and fuels annually.

The quantity and value of oil and by-products that could be recovered annually by liquefaction and upgrading of these wastes are estimated in Table 1. Byproducts include carbon black and steel wire from the tires, and aluminum foil derived from labels and lids on plastic containers.

Table 1. Quantities and value of potential products from waste polymers

Waste polymer	Tons/year	Oil (barrels/yr)	By-product (tons/yr)	Value (\$/yr)
Plastics	22 million	110 million	Al foil (110,000)	22,000,000
Tires	2.8 million	8.4 million	Carbon black (840,000)	168,000,000
			Steel wire (280,000)	14,000,000
Waste oil		30 million		
Total Oil		148 million		2,960,000,000
Total Potential	\$3,186,000,000			

German feedstock recycling industry

Germany currently has the most aggressive recycling program in the world. The development of the German recycling industry has been in response to very restrictive legislation that requires 80% of all consumer packaging materials to be recovered and 80% of all materials recovered to be recycled. The response of German industry to this law has been the creation of the Duales System Deutschland or DSD. Member companies of the DSD place a small surcharge (roughly a penny) on every container they sell. The money collected (~4 billion DM) is used to subsidize companies that collect, separate, prepare and recycle waste packaging material. DSD supports processing plants that convert the waste plastic into oil, olefins, synthesis gas, and reducing gases for production of steel in blast furnaces. The processing plant closest to the technology discussed here is the liquefaction plant of Koheöl-Anlage Bottrop, GmbH (KAB), which is currently liquefying 80,000 tons of DSD waste plastic feedstock per year. Members of the committee had many valuable interactions with representatives of the DSD and their contractors and a complete description of their operations is given in the full feasibility study report. (1)

Research Summary

Recent research in the U.S. has been summarized in several conference proceedings volumes. (5-7) Much research and development has also taken place in Germany. Some of the results that are most pertinent for demonstration plant development are given below.

<u>Plastic liquefaction and coprocessing:</u> The liquefaction of commingled waste plastic typically yields 80-90% oil, 5-10% gas, and 5-10% solid residue. Solid acid catalysts and metal-promoted solid acid catalysts improve oil yields and oil quality. At temperatures above 440 °C, thermal and catalytic oil yields are comparable but oil products of higher quality are produced catalytically. No solvent is required but good results have been with mixtures of waste oil and plastic. The reactions can be carried out at low hydrogen pressures (~100-500 psig) and with low hydrogen consumption (~1%).

Coprocessing of plastic with coal and resid yields the best results when using catalysts with both hydrogenation and hydrocracking functions, such as metal-promoted SiO₂-Al₂O₃ or mixtures of metal hydrogenation catalysts with HZSM-5. Oil yields of 40-70% and total conversions of 70-90% have been obtained. High hydrogen pressures and a solvent with some aromatic character, such as petroleum resid, are required.

Rubber liquefaction: Crumb rubber is readily liquefied at 400 °C under low hydrogen pressures (~100-500 psig), yielding 50-60% oil, 5-10% gas, and 30-40% carbon black. Experiments on the coprocessing of tire rubber with coal indicate that rubber converts to oil in the same manner as it does when coal is not present. High hydrogen pressures and a hydrogenation catalyst, such as nanoscale iron or molybdenum sulfide, are required for coprocessing of rubber and coal. Because of their relatively high content of carbon black (~30%) and wire (~10%), the feasibility committee concluded that the best approach for tires is to pyrolyse them and hydrotreat the pyrolysis oil, either alone or in mixtures with coal and/or plastic. The carbon black and wire can then be easily separated as byproducts of the process. Activation of the carbon black yields a carbon product with a surface area of several hundred m²/g.

Plant Design

A modular design was chosen for the demonstration plant. The three principal modules for the base design and their functions are described below.

- (1) <u>Tire module</u> The tires are shredded and pyrolysed at $\sim 600-700$ °C. The pyrolysis oil from tires is hydrotreated in the upgrading module. Steel wire is magnetically separated from the solid residue to be sold as scrap steel, while the carbon black is activated by partial oxidation.
- (2) Waste plastic module Melting/depolymerization (M/D) of the plastic is carried out at a moderate temperature (~380 °C). Light oils are condensed from the volatiles leaving this reactor and HCl is removed by scrubbing. Aluminum foil and other inerts sink to the bottom of the M/D reactor and are removed. Liquid product from the reactor is hydrocracked at 440-450 °C, either catalytically or thermally, and the total oil product is transferred to the upgrading module for hydroprocessing. Replacement of the M/D reactor by a pyrolysis reactor is another option.
- (3) <u>Upgrading module</u> Catalytic upgrading of the liquid products from modules 1 and 2 occurs in a slurry phase reactor using a dispersed, nanoscale, iron-based catalyst and distillation of the upgraded product is carried out.

A modular approach allows developers to choose the modules that best suit their needs. For example, if tire recycling is the main objective, modules 1 and 3 are needed. If waste plastic is the primary interest, modules 2 and 3 are required. However, module 2 alone produces relatively high quality oil that might meet the requirements of some developers. Several modular combinations are considered in the economic analysis.

Tires entering the pyrolysis reactor may be replaced by coal, while petroleum resid and waste oil may replace any of the liquids entering the upgrading module.

Economic analysis: Independent economic analyses for the plant were carried out by Harvey Schindler of Burns & Roe Services Corp. and by Mahmoud El-Halwagi and Mark Shelley of Auburn University and the CFFLS. (1) The uncertainty in the results of the economic analysis is considered to be fairly large (\pm 30%) for the following reasons:

- (1) The small size of the plant (200 tons/day of plastics and 100 tons/day of tires) necessitated scaling down the cost of much larger units.
- (2) There were wide variations in equipment cost quotes from different manufacturers.
- (3) The committee identified several unanswered research questions related to plant design.

The economic analysis was first carried out for the base case with the three modules described above processing 200 tons per day of waste plastic and 100 tons per day of tires (10,000 tires per day). Some of the additional cases considered included: (1) a plant liquefying 300 tons per day of plastics with no tires; (2) a plant liquefying 300 tons per day of tires (30,000 tires per day) with no plastics; (3) the replacement of up to 20% of the tires by coal; (4) the replacement of up to 20% of the waste plastic by petroleum resid or waste oil; and (5) the effect on the economics of increasing the size of the base case plant by a factor or either 2 or 5. A summary of the results for these plant development scenarios is presented Table 2. Revenues and returns on investment

Table 2. Summary of estimated capital costs and ROI for various plant scenarios.

Case	Capital cost (\$MM)	ROI (%), \$20/barrel	ROI (%), \$25/barrel
1. Base case: 200 t/d of plastic, 100 t/d tires.	55.2	8-16	12-20
2. 300 t/d of plastic, plastic + upgrading	55.5	10-15	15-20
module.			
3. 300 t/d of plastic, plastic module alone.	47.5	12-18	18-24
4. 300 t/d of tires, tire + upgrading module.	40.2	5-23	7-26
5. Case 4 with carbon priced at \$500/ton.	40.2	25-43	28-46
6. Base case with 20% waste oil/resid or	55.2	6-14	
20% coal replacing plastic or tires.			
7. Case 2 with 20 % waste oil/resid	55.5	9-14	
replacing plastic.			
8. Case 4 with 20% coal replacing tires.	40.2	(-1)-13	
9. Case 1 for a 600 t/d plant.	89.7	13-23	
10. Case 1 for a 1500 t/d plant.	170.3	21-35	

(ROI) are calculated assuming product values of \$20 or \$25 per barrel for oil, \$200 per ton for carbon, \$50 per ton for steel wire, and \$200 per ton for aluminum foil. The estimated capital cost for each case is given in column 2 and the range of returns on investment (ROI) is given in columns 3 and 4 for oil priced at \$20 and \$25 per barrel. The main factor determining the range of the ROI is the size of the tipping fees received by the plant for waste plastics and tires. In the table, the results are presented for plastic tipping fees from \$30 to \$60 per ton and for tire tipping fees ranging from -\$20 to \$75 per ton. \$30 to \$60 per ton of waste plastic is typical of current

landfill tipping fees in various parts of the U.S. For tires, a tipping fee of -\$20 per ton indicates that the developer has elected not to perform any tire shredding operations at the plant and is simply buying shredded tires from a waste tire processing company at a cost of \$20 per ton, while a tipping fee of \$75 per ton would be what the developer could expect to receive if tires are shredded at the plant. ROI results for the base case and for a 600 t/d plant are shown in Figure 1.

The results are encouraging. For the base case (200 tons/day of waste plastic, 100 tons/day of tires), the predicted ROI with oil priced at \$20 per barrel is 8-16%. For a plant processing plastic alone, ROIs up to 18% are predicted, while a plant processing tires alone could achieve a ROI up to 23%. Higher oil or carbon prices improve the ROI considerably.

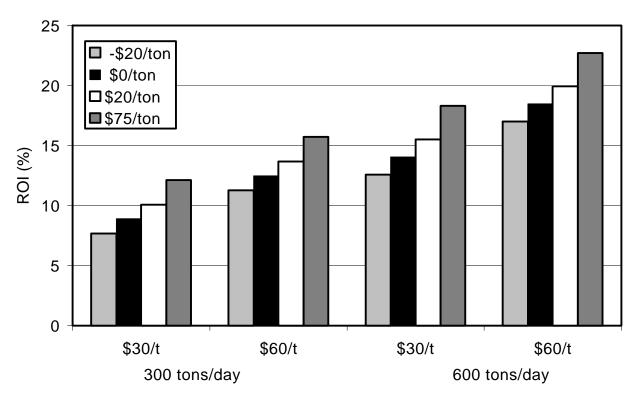


Figure 2. ROI for the base case as a function of tipping fees for plastic (x-axis) and tires (box) for two plant sizes.

If plastic and tires are replaced by waste oil/resid or coal (cases 5-7), the ROI decreases because of the loss of tipping fees. However, the decreases predicted are moderate. If 20% of the waste plastic feedstock is replaced by waste oil/resid, the decrease in ROI is expected to be only 1-2%, while replacement of 20% of the tire feedstock by coal will lead to ROI decreases of 2-10%.

Also included in the table are the results for several cases (cases 8-10) which achieve significantly higher ROIs. For example, if case 4 (300 tons of tires per day, no plastic) is considered with a higher value for the activated carbon product (\$500/ton or 25¢/lb.), the ROI is predicted to be 25-46%. Larger plants are also more profitable: a 600 ton/day plant could achieve ROI levels of 13-23%, while a 1500 ton per day plant could yield a ROI of 21-35%.

Future Program

The results of the current feasibility study indicate that a new industry can be developed to convert waste materials and fossil energy resources into valuable hydrocarbon fuels and byproducts. It is considered significant that the economic analysis predicts, for the first time, that a direct liquefaction process can be profitable and return a reasonable ROI at current oil prices, even at the demonstration plant stage.

As noted earlier, a significant resource is wasted by disposal of plastics and tires in landfills. However, plastics and tires constitute only about 12-14% by weight of all the waste materials that are landfilled. Other materials, such as paper wastes, yard wastes, wood wastes, biomass, and automobile shredder residue, are also largely hydrocarbon-based and constitute a much larger percentage (50-60%) of landfill mass. It therefore seems logical to investigate the conversion of these materials into valuable hydrocarbon fuels or chemicals in future research. Ideally, it would be advantageous to incorporate the conversion of these waste materials with the conversion of coal, possibly within the setting of an integrated gasification combined-cycle (IGCC) power plant. Pyrolysis, gasification, syngas reaction, activation, upgrading and hydroprocessing should all be investigated to achieve the most efficient conversion of different types of waste materials into valuable hydrocarbon products. The Feasibility Study Committee strongly recommends that the U.S. Department of Energy embark on such a program.

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